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Thermophysical Properties of Pure Refrigerants and Refrigerant Mixtures by Dynamic Light Scattering (DLS)

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ABSTRACT

The present paper summarizes the activities at LTT-Erlangen on the field of thermophysical properties research for pure refrigerants and refrigerant mixtures. For the characterization of their thermophysical properties dynamic light scattering (DLS) has been intensively applied as a key tool during the past two decades. Beside a representation of the essential features of DLS for the determination of thermophysical properties, the present paper gives a complete compilation of all experimental data obtained until now at LTT-Erlangen for thermal diffusivity, speed of sound, kinematic viscosity, and surface tension of pure refrigerants and refrigerant mixtures under saturation conditions. For refrigerant mixtures of technical interest the paper includes a review of literature data and deals with a simple prediction method based on the properties of the pure components expressed as functions of the reduced temperature. This calculation scheme was found to be applicable for the prediction of both equilibrium properties and transport properties of multicomponent refrigerant mixtures with sufficient high accuracy.

1. INTRODUCTION

In the course of the last twenty years, dynamic light scattering (DLS) has been used with increasing effort and attention at LTT-Erlangen for the measurement of thermophysical properties of pure refrigerants and refrigerant mixtures. First research activities for refrigerants by DLS at LTT-Erlangen were motivated by the prohibition of chlorinated hydrocarbons (CFCs) based on the Montreal Protocol in 1987. The major task of these activities was the routine application of light scattering from bulk fluids on a molecular level to rapidly provide comprehensive data sets for the thermal diffusivity and speed of sound under saturation conditions of some partially halogenated chlorofluorocarbons (HCFCs) considered as replacement fluids in a transient period and of chlorine-free hydrofluorocarbons (HFCs) evolved as long-term alternatives (Kraft and Leipertz, 1994a, 1994b, 1994c, 1996; Kraft, 1995). In a second step, by applying the DLS-technique to fluid surfaces, or in a more general formulation to phase boundaries, also called surface light scattering (SLS), the kinematic viscosity and surface tension could be measured for numerous pure refrigerants (Fröba *et al.*, 1999a, 2000, 2001a). In particular, this step was accompanied by a continuous improvement of the SLS-technique and the associated data evaluation procedure, see Fröba *et al.* (1999b) and Fröba and Leipertz (2001b, 2003a). Consequently, recalculated values for the liquid viscosity and surface tension of pure refrigerants and refrigerant mixtures under saturation conditions could be obtained with higher accuracy (Fröba, 2002). These data sets are here available for the first time in form of their correlations. Last research activities mainly concentrated on refrigerant mixtures. While mixtures of technical interest (Fröba *et al.*, 2001c, 2001d; Fröba and Leipertz, 2003b) have been investigated at given composition, with respect to the development and improvement of prediction models, during the latest years our special interest was also focused on the study of thermophysical properties of refrigerant mixtures in dependence on composition. Objects of these latter investigations which are not included here were a binary mixture of R143a + R125 (Fröba *et al.*, 2004a) and a quaternary mixture of R143a, R134a, R125, and R32 (Fröba *et al.*, 2006a).

In the following, however, only the investigations done until now at LTT-Erlangen by DLS for refrigerants of technical interest are compiled. Experimental data for the thermal diffusivity, speed of sound, kinematic viscosity, and surface tension are summarized for practical use in form of uniform correlations for the pure refrigerants R123, R142b, R23, R32, R125, R143a, R134a, R152a, R365mfc, and R227ea as well as for the refrigerant mixtures R507

(50 % wt. R143a / 50 % wt. R125), R404A (52 % wt. R143a / 44 % wt. R125 / 4 % wt. R134a), R410A (50 % wt. R125 / 50 % wt. R32), R407C (52 % wt. R134a / 25 % wt. R125 / 23 % wt. R32), and a newly developed azeotropic blend consisting of 65 % wt. R365mfc and 35 % wt. Galden® HT 55 (perfluoropolyether).

2. METHOD AND EXPERIMENTAL SETUP

The main advantage of the DLS-technique is based on the determination of thermophysical properties in macroscopic thermodynamic equilibrium. The method allows an absolute determination of a wide range of thermophysical properties with high accuracy using a basically identical experimental setup, e.g., thermal diffusivity, mutual diffusivity, speed of sound, sound attenuation, Landau-Placzek ratio, kinematic and dynamic viscosity, and surface tension (Leipertz and Fröba, 2005). In the following only some general aspects of the underlying theory of DLS from bulk fluids and the application of this method to fluid surfaces are remarked. For a detailed and comprehensive description the reader is referred to the specialized literature (Chu, 1991; Langewin, 1992).

In pure fluids, the thermal diffusivity can be found from the linewidth of the Rayleigh component of the spectrum of scattered light, which arises from entropy fluctuations. In binary fluid mixtures, the thermal diffusivity and mutual diffusivity can basically be determined simultaneously from the linewidth of the Rayleigh line, governed by microscopic fluctuations of temperature and concentration. Whether it is possible to determine signals simultaneously from concentration and temperature fluctuations is mainly governed by the relative difference of the refraction indices of the mixture components and their concentration. For the refrigerant mixtures of this work the refractive indices of the pure components have comparable values so that from the Rayleigh component of scattered light a signal from only temperature fluctuations associated with thermal diffusivity can be resolved. Additionally, information about sound velocity and sound attenuation can be obtained from the Brillouin lines of the spectrum, which are shifted in frequency with respect to the incident light and which are caused by pressure fluctuations. Information about surface tension and kinematic viscosity can be derived from light scattering by surface waves, which are caused by the thermal movement of molecules resulting in so-called “ripples”.

The optical and electro-optical components of the experimental setup used for the determination of sound speed and thermal diffusivity are shown - in a top view - on the left side of Fig. 1. For performing light scattering from bulk fluids, the scattering volume, determined by the intersection of the incident beam and the axis of observation (dashed line), is located in the middle of the vessel. The arrangement of the scattering geometry which enables scattering by surface waves is shown schematically - in a front view - on the right side of Fig. 1. In this case, the optical path has to be aligned in a way that the laser beam and the direction of detection intersect on the liquid-vapor interface in the measurement cell. The basic modification of the setup on the left side of Fig. 1 for carrying out surface light scattering experiments is the mounting of the pressure vessel in a vertical position. As light source, an argon ion laser ($\lambda_0 = 488$ nm) or optionally a frequency doubled continuous wave Nd:YVO₄ laser ($\lambda_0 = 532$ nm) were used. The laser power was up to 300 mW when working far away from the critical point, and only a few mW in the critical region. For large scattering intensities, scattered reference light from the cell windows alone is not sufficient to achieve heterodyne conditions; here, an additional reference beam is added. For this purpose, part of the

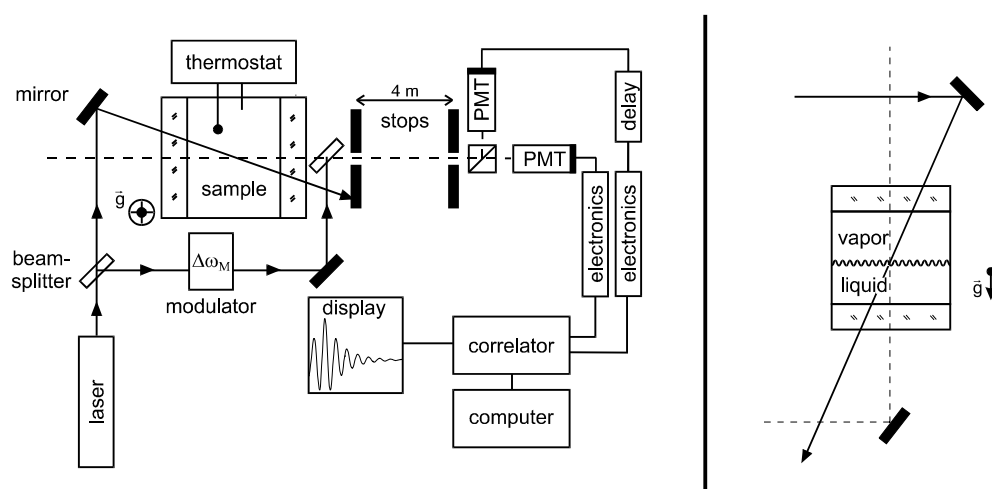


Figure 1: Experimental setup: optical and electronic arrangement.

incident laser light is split by a glass plate and superimposed with the scattered light behind the sample cell. For the determination of sound speed, a reference beam shifted in frequency by an opto-acoustic modulator was added to the scattered light. The time-dependent intensity of the scattered light is detected by two photomultiplier tubes (PMT's) operated in cross-correlation in order to suppress after-pulsing effects. The signals are amplified, discriminated, and fed to a digital correlator. The temperature of the sample cell was regulated through resistance heating and measured by calibrated 25- Ω or 100- Ω platinum resistance probes with an uncertainty of ± 0.015 K. The temperature stability was better than ± 0.002 K during one experimental run. For each temperature point, typically six measurements at different angles of incidence were performed, where the laser was irradiated from either side with respect to the axis of observation in order to check for a possible misalignment. A more detailed description of the experimental setups and the measurement procedures can be found in Fröba *et al.* (1999b) and Fröba and Leipertz (2003a).

3. RESULTS AND DISCUSSION

Different correlation schemes have been applied in the original works to represent the experimental data from DLS at saturation conditions, see, e.g., Kraft and Leipertz (1994b), Kraft and Leipertz (1996), and Fröba *et al.* (2001e). In the present summary for practical use an uniform data correlation has been performed for all pure refrigerants and refrigerant mixtures investigated until now at LTT-Erlangen.

For both phases, the experimental data for the thermal diffusivity as well as for the sound speed can be represented by the sum of a polynomial and an additional term, which accounts for the description of the critical behavior, according to

$$y = \sum_{i=0}^2 y_i (T / K)^i + \frac{y_3}{(T - T^*) / K} . \quad (1)$$

For the boiling and condensation curves, the coefficients of Eq. (1) as well as its range of obedience are summarized in Table 1. Notice that in the data fit according to Eq. (1), T^* is an additional fit parameter. Each of these correlations represents the data within experimental uncertainty. Except for R142b and R125 and for some data near the critical point, overall uncertainties of ± 1 % for the thermal diffusivity and of ± 0.5 % for the sound speed can be estimated. For measurements in the vicinity of the critical point, the uncertainty of both thermal diffusivity and sound speed increased to ± 2 % and ± 1 %, respectively, which can be attributed to the more difficult experimental accessibility of the critical region. Also listed in Table 1 is the root mean square deviation of the measured data from Eq. (1).

While a simple or modified Andrade-type equation may well represent the dynamic viscosity not too close to the critical point - and some authors have simply adopted this approach for the kinematic viscosity - this type of equation fails to reasonably represent the liquid kinematic viscosity for all refrigerants for the whole temperature range. Thus, we have chosen an empirical polynomial approach,

$$\nu = \sum_{i=0}^4 \nu_i (T / K)^i , \quad (2)$$

to represent the experimental viscosity data. The experimental data for the surface tension can well be represented by a modified van der Waals-type surface tension equation of the form

$$\sigma = \sigma_0 (1 - T / T_C)^{1.26} \left[1 + \sigma_1 (1 - T / T_C)^{0.5} + \sigma_2 (1 - T / T_C) \right] , \quad (3)$$

where T and T_C denote the temperature and the critical temperature, respectively. In Eq. (3), σ_0 , σ_1 , and σ_2 are fit parameters. The coefficients of Eqs. (2) and (3) are given in Table 2. Here, also the standard deviations of the experimental data relative to those calculated are listed. The critical temperatures given in Table 2 were either adopted from literature, theoretically calculated, or experimentally determined by the observation of the vanishing meniscus between the liquid and vapor phase when approaching the critical point. The total uncertainty of the data for the liquid kinematic viscosity and surface tension can be estimated to be less than ± 2 % and ± 1.2 %, respectively. A more detailed discussion regarding the accuracy achievable for the liquid kinematic viscosity and surface tension from SLS is given by Fröba and Leipertz (2003a) and Fröba (2002).

In order to predict the mixture data for thermal diffusivity and sound velocity of R507, R404A, R410A, and R407C on basis of the pure component data within the range investigated, several different approaches have been tried using, e.g., simple weighting by the mass fractions or the mole fractions of the pure components. In all these cases, remarkable deviations appeared in the vicinity of the critical point when plotting the data against temperature. Involving, however, properties based on the reduced temperature, excellent agreement was found between the measured data and the predicted values when weighting with the mass fractions. The agreement was not this good

Table 1: Coefficients of Eq. (1).

y_i	Thermal Diffusivity a Liquid Phase	y_i ($10^{-8} \text{ m}^2 \cdot \text{s}^{-1}$) Vapor Phase	Sound Speed c_s Liquid Phase	y_i ($\text{m} \cdot \text{s}^{-1}$) Vapor Phase
R142b (1-Chloro -1,1-Difluoroethane) Original Report: Kraft and Leipertz (1994b); Kraft (1995)				
y_0	13.0165	134.3704	1502.31	272.05
y_1	-0.0296401	-0.3268713	-1.86427	-0.30008
y_2	1.598×10^{-5}	—	-0.0035209	—
y_3	22.977	—	602.5	2437.6
T^* (K)	415.3467	—	418.233	455.87
rms (%)	0.89	2.95	0.27	0.24
T -range (K)	278.7 – 406.4	368.7 – 403.9	288.7 – 406.4	373.8 – 405.3
R32 (Difluoromethane) Original Report: Kraft and Leipertz (1994b); Kraft (1995)				
y_0	8.50717	109.46727	634.01	-317.53
y_1	0.0298342	-0.3103268	5.1381	3.90744
y_2	-10.84513×10^{-5}	—	-0.0180552	-0.0072072
y_3	71.275	0.3758	504.4	62.5
T^* (K)	364.295	352.4747	357.794	353.522
rms (%)	0.57	0.81	0.19	0.09
T -range (K)	278.6 – 350.5	328.5 – 350.5	293.7 – 350.8	317.6 – 350.9
R125 (Pentafluoroethane) Original Report: Kraft and Leipertz (1994a)				
y_0	7.2879	106.6209	2077.27	133.04
y_1	-0.0103684	-0.312481	-6.98936	0.02325
y_2	—	—	0.00410703	—
y_3	37.902	1.4144	1735.67	1774
T^* (K)	349.9388	341.5022	356.094	367.85
rms (%)	2.87	2.55	0.27	1.75
T -range (K)	292.2 – 337.8	306.5 – 338.2	293.1 – 338.3	306.5 – 338.2
R143a (1,1,1-Trifluoroethane) Original Report: Fröba <i>et al.</i> (2001e)				
y_0	28.9074	121.2187	1255.44	-214.97
y_1	-0.1395383	-0.3484418	-0.71304	3.0039
y_2	19.99114×10^{-5}	—	-0.0071096	-0.0059994
y_3	41.5167	2.1416	666.2	24.01
T^* (K)	355.2227	349.757	355.5	347.828
rms (%)	0.78	0.84	0.25	0.11
T -range (K)	273.1 – 345.1	318.12 – 345.1	288.1 – 345.0	313.1 – 345.6
R134a (1,1,1,2-Tetrafluoroethane) Original Report: Kraft and Leipertz (1996)				
y_0	13.14178	120.86291	1347.19	-548.57
y_1	-0.0337004	-0.3213981	-1.15173	4.6573
y_2	2.10795×10^{-5}	—	-0.0055021	-0.00783204
y_3	24.4835	2.9048	615.44	16.1
T^* (K)	381.0286	378.5999	383.41	375.292
rms (%)	1.67	0.74	0.18	0.10
T -range (K)	294.9 – 373.9	355.7 – 373.8	294.9 – 373.8	333.15 – 373.8
R152a (1,1-Difluoroethane) Original Report: Kraft and Leipertz (1994c)				
y_0	20.1291	136.47735	1868.89	456.06
y_1	-0.0664327	-0.3502791	-3.2768	-0.82165
y_2	7.1423×10^{-5}	—	-0.00257415	—
y_3	53.398	10.171	1702.6	140
T^* (K)	397.2231	397.969	401.837	392.27
rms (%)	0.41	0.43	0.26	0.03
T -range (K)	278.7 – 385.8	360.1 – 385.8	278.7 – 385.8	360.1 – 385.8
R227ea (1,1,1,2,3,3,3-Heptafluoropropane) Original Report: Fröba <i>et al.</i> (2005)				
y_0	18.24094	119.649243	1581.27	-1116.87
y_1	-0.0801275	-0.31647457	-3.82095	7.604
y_2	10.60593×10^{-5}	—	-0.000134082	-0.0118747
y_3	17.7732	5.19839	1151.445	8.88
T^* (K)	380.6475	380.3553	390.748	377.428
rms (%)	0.95	0.58	0.29	0.11
T -range (K)	293.2 – 374.7	345.7 – 374.7	293.2 – 374.0	345.7 – 374.0

Table 1 (Continued)

y_i	Thermal Diffusivity a Liquid Phase	y_i ($10^{-8} \text{ m}^2 \cdot \text{s}^{-1}$) Vapor Phase	Sound Speed c_s Liquid Phase	y_i ($\text{m} \cdot \text{s}^{-1}$) Vapor Phase
R365mfc (1,1,1,3,3-Pentafluorobutane) Original Report: Fröba <i>et al.</i> (2004b)				
y_0	11.1672	152.5488	2032.37	-1081.24
y_1	-0.029097	-0.3282257	-4.89882	6.28303
y_2	2.60647×10^{-5}	—	0.00211016	-0.00820422
y_3	18.487	5.85	6698.6	10.14
T^* (K)	466.7	463.5445	502.378	461.64
rms (%)	0.39	0.82	0.17	0.38
T -range (K)	298.2 – 458.2	428.2 – 458.2	298.2 – 459.2	428.2 – 459.2
R507 (R125 / R143a) Original Report: Fröba <i>et al.</i> (2001c)				
y_0	18.30268	107.88065	1074.93	-510.92
y_1	-0.0715551	-0.3087015	0.166	4.691
y_2	8.2522×10^{-5}	—	-0.0083805	-0.0085025
y_3	22.9437	23.571	537.66	24.18
T^* (K)	350.4519	358.698	352.764	346.216
rms (%)	0.82	0.41	0.32	0.13
T -range (K)	293.1 – 343.1	319.1 – 343.1	293.0 – 342.6	315.1 – 343.1
R404A (R125 / R143a / R134a) Original Report: Fröba <i>et al.</i> (2001c)				
y_0	21.8999	117.18934	887.79	-711.5
y_1	-0.093882	-0.3382481	1.34198	5.9663
y_2	11.82656×10^{-5}	—	-0.0101673	-0.0105148
y_3	25.9746	0.57983	554	9.78
T^* (K)	352.3386	345.9125	354.66	345.96
rms (%)	0.42	0.83	0.16	0.07
T -range (K)	303.1 – 344.2	318.1 – 344.2	303.1 – 344.2	318.1 – 344.2
R410A (R125 / R32) Original Report: Fröba and Leipertz (2003b)				
y_0	7.55763	114.41403	50.11	-637.82
y_1	0.0130127	-0.3318855	7.88183	5.65123
y_2	-6.9666×10^{-5}	—	-0.022108	-0.0099897
y_3	31.2096	0.07827	153.15	32.55
T^* (K)	352.4123	343.8875	347.093	345.886
rms (%)	0.36	0.65	0.12	0.11
T -range (K)	298.1 – 343.2	315.6 – 343.1	298.1 – 343.2	315.6 – 343.1
R407C (R125 / R32 / R134a) Original Report: Fröba and Leipertz (2003b)				
y_0	2.85065	—	1125.34	—
y_1	0.0350417	—	0.39816	—
y_2	-9.52585×10^{-5}	—	-0.00859288	—
y_3	24.6652	—	700.8	—
T^* (K)	367.909	—	371.527	—
rms (%)	0.55	—	0.18	—
T -range (K)	293.2 – 359.1	—	293.1 – 359.1	—
R365mfc / Galden HT 55 (Perfluoropolyether) Original Report: Fröba <i>et al.</i> (2006b)				
y_0	13.1412	123.1588	1590.04	-187.63
y_1	-0.0454664	-0.2654692	-2.9979	1.80617
y_2	4.9631×10^{-5}	—	-0.00060745	-0.00259925
y_3	10.1133	54.532	1228.6	1520
T^* (K)	453.3985	466.2285	469.285	487.39
rms (%)	0.91	1.08	0.24	0.11
T -range (K)	298.2 – 447.2	418.2 – 449.2	293.2 – 448.2	373.2 – 449.2

using mole fractions. Thus, the properties of the mixture Y_M under saturation conditions have been predicted according to

$$Y_M(T_R) = \sum_j w_j Y_j(T_R) \quad (4)$$

where w_j and Y_j are the mass fraction and the property at the reduced temperature $T_R = T/T_C$ of component j , respectively. In Fig. 2 the experimental results for the thermal diffusivity and sound velocity of R507, R404A, R410A, and R407C are compared with this simple prediction method. To this end, the pure component data for thermal

Table 2: Coefficients of Eq. (2) and Eq. (3).

Eq. (2)	ν'_0 (mm ² ·s ⁻¹)	$\nu'_1 \times 10^2$ (mm ² ·s ⁻¹)	$\nu'_2 \times 10^5$ (mm ² ·s ⁻¹)	$\nu'_3 \times 10^8$ (mm ² ·s ⁻¹)	$\nu'_4 \times 10^{10}$ (mm ² ·s ⁻¹)	T-range (K)	rms (%)
Eq. (3)	—	σ_0 (mN·m ⁻¹)	σ_1	σ_2	T_c (K)	T-range (K)	rms (%)
R123 (2,2-Dichloro-1,1,1-trifluoroethane) Original Report: Fröba (2002)							
Eq. (2)	6.54962	-5.068267	13.78739	-12.85555	—	253 – 373	0.80
Eq. (3)	—	51.039	0.666	-0.757	456.831	253 – 373	0.31
R23 (Trifluoromethane) Original Report: Fröba (2002)							
Eq. (2)	2.28673	-2.222899	7.811277	-9.580752	—	233 – 293	0.61
Eq. (3)	—	59.808	0.321	-0.590	298.98	233 – T_c	0.83
R32 (Difluoromethane) Original Report: Fröba (2002)							
Eq. (2)	2.47709	-2.102963	6.419838	-6.772865	—	233 – 343	1.15
Eq. (3)	—	70.362	0.415	-0.686	351.35	233 – T_c	1.45
R125 (Pentafluoroethane) Original Report: Fröba (2002)							
Eq. (2)	3.33135	-2.879739	8.884091	-9.524125	—	233 – 333	0.90
Eq. (3)	—	51.112	0.455	-0.667	339.33	233 – T_c	2.26
R143a (1,1,1-Trifluoroethane) Original Report: Fröba (2002)							
Eq. (2)	1.38133	-0.913776	2.271086	-2.079078	—	253 – 333	0.59
Eq. (3)	—	51.612	0.362	-0.505	345.857	253 – T_c	0.74
R134a (1,1,1,2-Tetrafluoroethane) Original Report: Fröba (2002)							
Eq. (2)	5.03354	-4.235156	12.46866	-12.55540	—	243 – 363	0.81
Eq. (3)	—	54.884	0.555	-0.712	374.18	243 – T_c	2.81
R152a (1,1-Difluoroethane) Original Report: Fröba (2002)							
Eq. (2)	3.42909	-2.653383	7.353200	-7.064462	—	243 – 373	0.42
Eq. (3)	—	66.943	-0.155	0.004	386.41	243 – T_c	1.20
R227ea (1,1,1,2,3,3,3-Heptafluoropropane) Original Report: Fröba <i>et al.</i> (2005)							
Eq. (2)	4.13024	-3.286066	9.294903	-9.133307	—	253 – 372	0.56
Eq. (3)	—	52.601	-0.088	0.028	374.90	253 – T_c	3.51
R365mfc (1,1,1,3,3-Pentafluorobutane) Original Report: Fröba <i>et al.</i> (2004b)							
Eq. (2)	45.3073	-52.14768	229.8266	-454.7764	33.91437	253 – 373	0.65
Eq. (3)	—	71.176	-0.5836	0.3881	460	253 – 373	0.39
R507 (R125 / R143a) Original Report: Fröba (2002)							
Eq. (2)	2.23108	-1.747084	4.998914	-5.073787	—	243 – 333	0.19
Eq. (3)	—	44.594	1.033	-1.272	344.06	243 – T_c	1.65
R404A (R125 / R143a / R134a) Original Report: Fröba (2002)							
Eq. (2)	2.80712	-2.320866	6.909354	-7.193090	—	243 – 333	0.29
Eq. (3)	—	49.757	0.552	-0.783	345.33	243 – T_c	0.67
R410A (R125 / R32) Original Report: Fröba and Leipertz (2003b)							
Eq. (2)	2.51768	-2.104374	6.360746	-6.725671	—	243 – 333	0.69
Eq. (3)	—	67.468	-0.051	-0.193	344.08	243 – T_c	0.76
R407C (R125 / R32 / R134a) Original Report: Fröba and Leipertz (2003b)							
Eq. (2)	2.65733	-2.018330	5.498332	-5.243336	—	243 – 333	0.51
Eq. (3)	—	48.206	1.049	-1.123	359.9	243 – T_c	3.82
R365mfc /Galden HT 55 (Perfluoropolyether) Original Report: Fröba <i>et al.</i> (2006b)							
Eq. (2)	25.4228	-26.32552	104.9700	-188.5389	12.78380	253 – 413	0.89
Eq. (3)	—	41.14	1.254	-1.507	450.7	253 – 413	0.88

diffusivity and sound velocity were taken from Kraft and Leipertz (1994a, 1994b, 1996), Kraft (1995), and Fröba *et al.* (2001a). As can be seen from the deviation plots, excepting the thermal diffusivity of R407C the predictions are very good with deviations typically smaller than 1 % for the sound speed and smaller than 2 to 5 % for the thermal diffusivity. Discrepancies between the predicted and experimental values for the thermal diffusivity of R407C in the liquid phase may be attributed to different curvatures for the pure components. Data for the thermal diffusivity of R134a in the liquid phase intersect those of R32. In the figures, also mixture data from reference databases (NIST, 2000; SOLVAY, 1999) and an experimental work (Hoffmann *et al.*, 1996) are included. For the latter, data for the liquid thermal conductivity λ have been converted to thermal diffusivity ($a = \lambda \cdot \rho^{-1} \cdot c_p^{-1}$) by using reference values for the specific heat at constant pressure c_p (Günther and Steimle, 1996) and density ρ (SOLVAY, 1999). For R507,

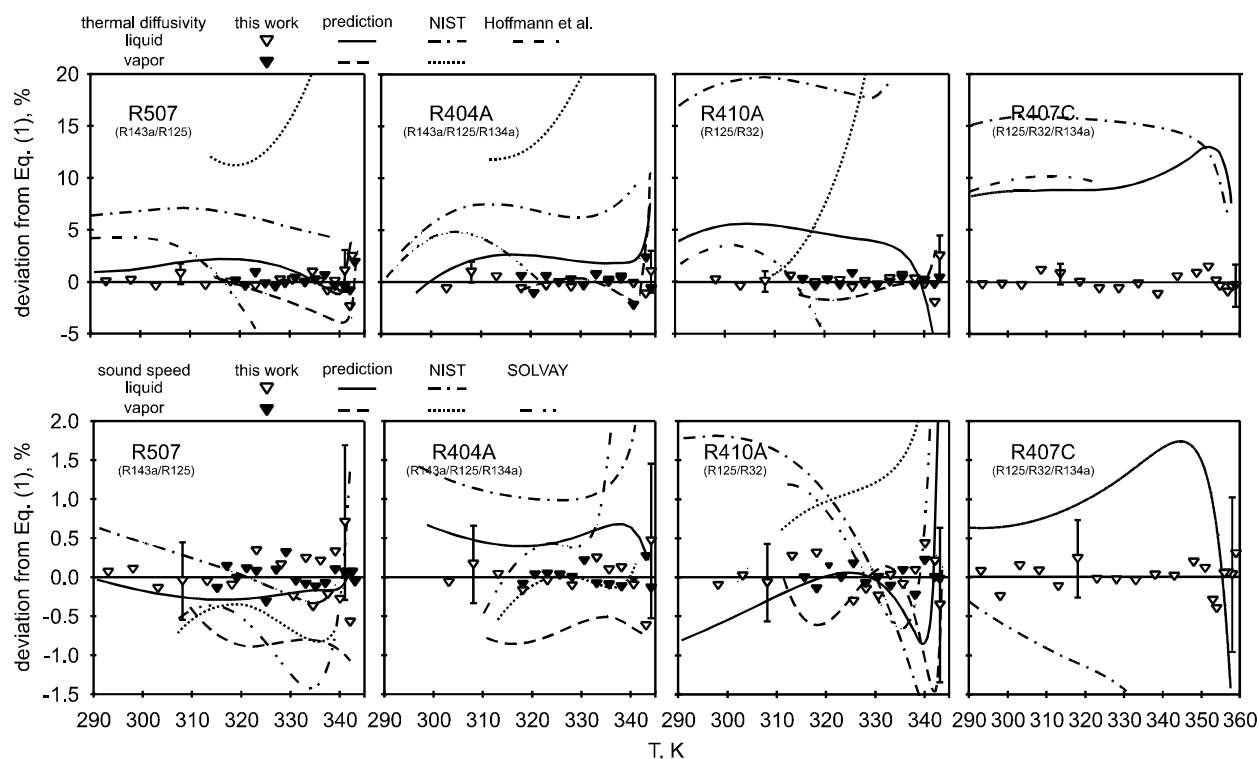


Figure 2: Data comparison for R507, R404A, R410A, and R407C in the liquid and vapor phases under saturation conditions. Top: thermal diffusivity; bottom: sound speed.

R404A, and R410A the thermal conductivity data of Hoffmann *et al.* (1996) deviate from the correlations in a range of about -5 to $+5$ %. These differences are within the combined uncertainties of the different methods, if one takes into account the uncertainty of the heat capacity data of less than 1 % for temperatures up to 313 K and less than 2 % for the temperature range between 313 to 323 K, according to Günther and Steimle (1996). In contrast, for R407C the deviations between the DLS measurements and the converted data are partly larger than 10 %, which exceeds the combined estimated uncertainties.

The mixture results for the liquid kinematic viscosity and surface tension of R507, R404A, R410A, and R407C under saturation conditions also follow quite well a mass-weighted calculation scheme according to Eq. (4). For a detailed discussion of these results in connection with the simple prediction scheme the reader is referred to Fröba *et al.* (2001d) and Fröba and Leipertz (2003b).

4. CONCLUSIONS

During the past two decades DLS has been applied successfully at LTT-Erlangen for the investigation of several thermophysical properties of refrigerants. The reason for the success of the method may be summarized as follows. Measurements may be performed in thermodynamic equilibrium, without an input of energy. Experiments are conducted in a non-contact mode, allowing access to regions of thermodynamic state which can hardly be probed by other techniques. Summarizing, for more than 10 pure refrigerants and refrigerant mixtures of technical interest an essential contribution to the improvement and verification of their data situation could be achieved.

REFERENCES

- Chu, B., 1991, *Laser Light Scattering*, Academic Press, New York.
- Fröba, A.P., Will, S., Leipertz, A., 1999a, Kinematic Viscosity and Surface Tension of Refrigerants from Surface Light Scattering, *Proc. 20th International Congress of Refrigeration IIR/IIF (Sydney)*, vol. II, paper 275.
- Fröba, A.P., Will, S., Leipertz, A., 1999b, Determination of Several Thermophysical Properties of Toluene Using a Single Experimental Setup, *Fluid Phase Equilib.*, vol. 161: p. 337-351.

- Fröba, A.P., Will, S., Leipertz, A., 2000, Saturated Liquid Viscosity and Surface Tension of Alternative Refrigerants, *Int. J. Thermophys.*, vol. 21: p. 1225-1253.
- Fröba, A.P., Will, S., Leipertz, A., 2001a, Thermophysical Properties of R143a (1,1,1-Trifluoroethane), *Int. J. Refrig.*, vol. 24: p. 734-743.
- Fröba, A.P., Leipertz, A., 2001b, Viscosity and Surface Tension of Saturated Toluene from Surface Light Scattering (SLS), *Int. J. Thermophys.*, vol. 22: p. 41-59.
- Fröba, A.P., Will, S., Leipertz, A., 2001c, Thermophysical Properties of Binary and Ternary Fluid Mixtures from Dynamic Light Scattering, *Int. J. Thermophys.*, vol. 22: p. 1349-1368.
- Fröba, A.P., Will, S., Leipertz, A., 2001d, Thermophysical Properties of the Refrigerant Mixtures R507, R410A, and R404A from Dynamic Light Scattering, *Proc. IIR Conference Thermophysical Properties and Transfer Processes of New Refrigerants (Paderborn)*, p. 14-21.
- Fröba, A.P., Will, S., Leipertz, A., 2001e, Thermal Diffusivity and Speed of Sound of the Refrigerant R143a (1,1,1-Trifluoroethane), *Int. J. Thermophys.*, vol. 22: p. 1021-1033.
- Fröba, A.P., 2002, *Simultane Bestimmung von Viskosität und Oberflächenspannung transparenter Fluide mittels Oberflächenlichtstreuung*, PhD Thesis, Universität Erlangen-Nürnberg.
- Fröba, A.P., Leipertz, A., 2003a, Accurate Determination of Liquid Viscosity and Surface Tension Using Surface Light Scattering (SLS): Toluene under Saturation Conditions between 260 K and 380 K, *Int. J. Thermophys.*, vol. 24: p. 895-921.
- Fröba, A.P., Leipertz, A., 2003b, Thermophysical Properties of the Refrigerant Mixtures R410A and R407C from Dynamic Light Scattering (DLS), *Int. J. Thermophys.*, vol. 24: p. 1185-1206.
- Fröba, A.P., Kremer, H., Leipertz, A., 2004a, Thermophysical Properties of Binary Mixtures of R125 + R143a in Comparison with a Simple Prediction Method, *Int. J. Thermophys.*, vol. 25: p. 1115-1133.
- Fröba, A.P., Krzeminski, K., Leipertz, A., 2004b, Thermophysical Properties of 1,1,1,3,3-Pentafluorobutane (R365mfc), *Int. J. Thermophys.*, vol. 25: p. 987-1004.
- Fröba, A.P., Botero, C., Leipertz, A., 2005, Thermal Diffusivity, Sound Speed, Viscosity, and Surface Tension of R227ea (1,1,1,2,3,3,3-Heptafluoropropane), submitted to *Int. J. Thermophys.*.
- Fröba, A.P., Botero, C., Leipertz, A., 2006a, Thermophysical Properties of a Quaternary Refrigerant Mixture: Comparison of Dynamic Light Scattering Measurements with a Simple Prediction Method, to be submitted to *Int. J. Thermophys.*.
- Fröba, A.P., Kremer, H., Botero, C., Leipertz, A., Flohr, F., Meurer, C., 2006b, Thermophysical Properties of a Refrigerant Mixture of R365mfc (1,1,1,3,3-Pentafluorobutane) and Galden HT 55 (Perfluoropolyether) to be submitted to *Int. J. Thermophys.*.
- Günther, D., Steimle, D., 1996, *Bestimmung der Transportgrößen von HFKW*, Bericht zum AiF-Forschungsvorhaben Nr. 10044B, Heft 3: Spezifische Wärmekapazität, Forschungsrat Kältetechnik e.V. (ed.), Frankfurt/M..
- Hoffmann, N., Spindler, K., Hahne, E., 1996, *Bestimmung der Transportgrößen von HFKW*, Bericht zum AiF-Forschungsvorhaben Nr. 10044B, Heft 2: Wärmeleitfähigkeit, Forschungsrat Kältetechnik e.V. (ed.), Frankfurt/M..
- Kraft, K., Leipertz, A., 1994a, Thermal Diffusivity and Ultrasonic Velocity of Saturated R125, *Int. J. Thermophys.*, vol. 15: p. 387-399.
- Kraft, K., Leipertz, A., 1994b, Thermal Diffusivity and Sound Velocity of Alternative Refrigerants by Dynamic Light Scattering, *Proc. International Conference CFCs, THE DAY AFTER Joint Meeting of IIR Commissions B1, B2, E1 and E2 (Padova)*, p. 435-442.
- Kraft, K., Leipertz, A., 1994c, Thermal Diffusivity and Ultrasonic Velocity of Sound of Saturated R152a, *Int. J. Thermophys.*, vol. 15: p. 791-802.
- Kraft, K., 1995, *Bestimmung von Schallgeschwindigkeit und Schalldämpfung transparenter Fluide mittels der Dynamischen Lichtstreuung*, PhD Thesis, Universität Erlangen-Nürnberg.
- Kraft, K., Leipertz, A., 1996, Thermal Diffusivity and Sound Velocity of Round-Robin R134a, *Fluid Phase Equilib.*, vol. 125: p. 245-255.
- Langewin, D., 1992, *Light Scattering by Liquid Surfaces and Complementary Techniques*, Marcel Dekker, New York.
- Leipertz, A., Fröba, A.P., 2005, Diffusion Measurements in Fluids by Dynamic Light Scattering, In: Heitjans, P., Kärger, J., *Diffusion in Condensed Matter – Methods, Materials, Models*, Springer, Berlin: p. 581-620.
- NIST, 2000, *Standard Reference Database 14*, Version 4.
- SOLVAY Fluor & Derivate, 1999, SOLKANE[®] Refrigerant Software, Version 2.0.